NATIONAL
URANIUM
TAILINGS
PROGRAM

Canada

LITERATURE REVIEW FOR THE NATIONAL URANIUM TAILINGS PROGRAM (NUTP) SEDIMENTATION STUDY

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A Report Prepared for:

The National Uranium Tailings Program
Canada Centre for Mineral and Energy Technology
Energy, Mines and Resources Canada
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National Uranium Tailings Program
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Attention: Roy D. John, Manager - Measurements

Gentlemen:

Re: Literature Review for National Uranium Tailings Program (NUTP) Sedimentation Study. Reference No. 15SQ.23241-4-1688

Monenco Consultants Limited is pleased to present 25 copies of the report entitled "Literature Review for the National Uranium Tailings Program (NUTP) Sedimentation Study".

This report addresses 17 questions relating to the removal of radionuclides from surface waters by sedimentation and remobilization of radionuclides from the sediment. Of the original 17 questions in the terms of reference, two questions:

- How do sedimentation rates of radionuclides vary? and
- What factors influence the sedimentation rates of radionuclides?

were combined into one question due to the lack of information.

As stated in our proposal, each question was addressed by:

- A discussion of literature pertinent to the question;
- A discussion of the sufficiency of the information available to answer the question; and

o The resolution of the question in terms of either a direct answer or the necessary types of work (e.g. laboratory experiments or field investigations) which needs to be conducted to establish the information necessary to satisfactorily answer the question.

During our review of the literature, Dr. R.J. Cornett of the Chalk River Nuclear Laboratories provided a prepublication manuscript of his recent work as well as much helpful information generally relating to radionuclides and sedimentation. Dr. J.A. Robbins provided his 1985 work dealing with sediment reworking and provided several unpublished manuscripts on this subject.

Yours very truly,

M.J. Riddle, P.Eng.
Manager
Environmental Division

EXECUTIVE SUMMARY

A review of the published literature and personal communication with experts in the field of natural radionuclides was conducted to address specific questions for the National Uranium Tailings Program sedimentation study. The 17 individual questions relating to the removal from surface water and the remobilization of radionuclides associated with uranium mining were categorized into five main groups:

- The radionuclides of concern;
- The forms taken by radionuclides entering the aquatic environment;
- The removal of radionuclides from surface waters;
- Sedimentation of radionuclides and dispersion within sediments; and
- Remobilization of radionuclides from sediment.

The radionuclides of major concern in the aquatic environment relative to the impacts of uranium mining and milling are:

- ^{226}Ra;
- ^{228}Ra (if the ore contains natural thorium);
- ^{210}Pb;
- ^{238}U, ^{234}U; and
- ^{230}Th and ^{232}Th.

There has been, to date, no definitive study of the form taken by radionuclides entering the aquatic environment from uranium milling tailings. However, based upon the known chemical conditions within the tailings, the chemical characteristics of the water flowing through or over the tailings, and the known aqueous chemistry of the radionuclides, it is possible to make hypotheses concerning the form of each radionuclide entering the aquatic environment.

The general mechanisms by which radionuclides are removed from surface waters can be described as:

- o chemical adsorption to organic and inorganic particulate material;
- o absorption by living particulate material;
- o incorporation into zooplankton fecal pellets;
- o precipitation scavenging; and
- o ion exchange.

The available information suggests that adsorption onto and ion exchange with living and nonliving particulate materials plays the largest role in removing radionuclides from surface waters.

Many publications address the use of ^{210}Pb in geochronological studies of sediments. This literature demonstrates a wide variety of sedimentation rates. The most recent literature (i.e. 1985) discusses the reasons for variation of the published rates. Whole-lake studies of radium uptake describe mechanisms of removal which may involve sedimentation as well as direct adsorption from the water column by plants. Comparatively little is known about sedimentation of other radionuclides in the freshwater environment.

Extensive and detailed laboratory microcosm studies have been carried out to define mathematically the mixing of radionuclide-labelled sediment by vertically oriented, deposit feeding macrobenthos (mainly tubificid worms and benthic amphipods).

Specified studies concerning the remobilization or release of radionuclides from sediment were not found during the literature survey; however, remobilization was sometimes implied in order to explain increased radionuclides concentrations in the water above sediments. The general chemistry of the radionuclides also demonstrates those conditions under which they have the potential to be released from the sediment.

Of the 17 individual questions relating to the removal of radionuclides from surface waters, eight were answered by the available literature: either the literature specifically addressed the question or the answer could be inferred from the literature. Three questions (concerning sedimentation rates) could only be addressed for one radionuclide (Pb-210). There was insufficient information in the literature to address six remaining questions.

These questions dealt with:

- the variations of ionic versus organically complexed radionuclides entering aquatic environments;
- the sedimentary history of areas affected by uranium milling operations;
- the use of sediment cores in these areas for sediment history determination and predictive modelling;
- the variations in sedimentation rates and the factors influencing these rates;
- the specific locations within these areas for sediment coring; and
- the potential remobilization mechanisms, their extent and occurrence.

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INTRODUCTION

PART 1 - INTRODUCTION

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1.0 INTRODUCTION

The National Uranium Tailings Program (NUTP) has as its mandate the formulation of a predictive mathematical model for the long-term fate of radionuclides introduced into the aquatic environment as the result of uranium mining and milling operations. Radionuclides entering the aquatic environment have the potential to become buried in sediments and thus be removed from the environment. To better understand the role of sedimentation in the removal of radionuclides from the aquatic environment, a series of questions relating to the sedimentation and remobilization of radionuclides in the aquatic environment was prepared by the NUTP staff.

The first objective in developing answers to these questions was to review current scientific literature to determine if the questions could be satisfactorily answered from existing information. To accomplish this objective, Monenco conducted a literature review using key words for entry into computerized data bases such as:

- Ocean Abstracts;
- Enviroline;
- Pollution Abstracts;
- Aquatic Science Abstracts;
- Aptic;
- Environmental Bibliography; and
- Aqualine.

Monenco's radionuclide literature files were also searched for pertinent publications. As well, personal contact was made with current researchers in this area.

As the review progressed, it was evident that the 17 questions posed by the NUTP could be broken down into the following categories:

- o The Radionuclides of Concern;
- o The Form Taken by the Radionuclides Entering the Aquatic Environment;
- o The Removal of Radionuclides from Surface Waters;
- o The Sedimentation of Radionuclides and Dispersion Within the Sediment; and
- o The Remobilization of Radionuclides from the Sediment.

The questions in each category are examined separately by first discussing the literature pertinent to the question. Following this is a discussion of the sufficiency of the information found in the literature review. Finally, the answer to the question is stated, or alternately, the work necessary to answer the question is discussed.

1.1 THE RADIONUCLIDES OF CONCERN

The question addressed is:

- o What radionuclides should be studied?

The published literature was reviewed for experts' opinions as to the potential hazard resulting from radionuclides released to the aquatic environment from uranium mining and milling.

1.2 THE FORM TAKEN BY THE RADIONUCLIDES ENTERING THE AQUATIC ENVIRONMENT

The questions addressed are:

- What proportions of the typically occurring radionuclides entering receiving waters from uranium tailings are in the ionic form as opposed to an organic complex?
- What variations occur in the ionic and organic complex proportions?
- What specific organic species are involved and what are the most likely organic chemical reactions taking place?

The published literature was searched for information concerning the leaching of radionuclides from mine tailings, the composition of mine water and the aqueous chemistry of the radionuclides of concern. The role of humic and fulvic acids in the formation of soluble complexes was also investigated in appreciation of their role in the formation of metal-organic complexes in the aquatic environment.

1.3 THE REMOVAL OF RADIONUCLIDES FROM SURFACE WATERS

The questions addressed are:

- What are the mechanisms for removal of radionuclides from surface waters?
- What roles do absorption, adsorption and precipitation play in the removal of radionuclides from surface waters?

- What roles do biological organisms play in the removal of radionuclides from surface waters?

- What is the relationship between the pelagic, profundal and littoral zones of a lake in the removal of radionuclides from surface waters by biological organisms?

The published literature was searched for information concerning the uptake of natural radionuclides by phytoplankton, zooplankton, aquatic vascular plants, attached algae, fish and benthic organisms. The main concern was to see if adsorption could be differentiated from absorption and which process played the greater role. Information was also sought concerning the removal of naturally occurring radionuclides by inorganic suspended particles alone.

1.4 SEDIMENTATION OF RADIONUCLIDES AND DISPERSION WITHIN SEDIMENTS

The questions addressed are:

- What is the sedimentation history for areas known to be affected by uranium milling operations?

- Can sediment cores collected from these areas be used to define a sedimentary history that will be useful in predictive modelling?

- What techniques should be used to obtain samples for historic analysis?

- What specific locations could be used to efficiently gain an understanding of past sedimentation rates?

- What are currently existing sedimentation rates of radionuclides?

- How do sedimentation rates of radionuclides vary and what factors influence the sedimentation rates?

- What effects do zoobenthic organisms have on the redistribution of radionuclides within active sediment layers?

The published literature was searched for information concerning sedimentation rates of radionuclides. As well, information concerning the geochronologies of areas known to be affected by uranium mining and milling were also sought. The patterns of sediment accumulation in lakes were reviewed with the intent of proposing techniques and general locations within lakes for obtaining sediment samples. The literature concerning bioturbation and its relation to the dispersal of radionuclides within the active sediment layer was also examined.

1.5 REMOBILIZATION OF RADIONUCLIDES FROM SEDIMENT

The question addressed is:

- What are the potential mechanisms for remobilization of radionuclides from sediment, their occurrence and extent of occurrence?

The published literature was searched for information concerning the conditions under which radionuclides were known to be released from the sediment or from particulate material with the potential to become part of the sediment.

1.6 FORMAT OF REPORT

Each of the above questions was addressed in the following format:

- o A discussion of literature pertinent to the question;
- o A discussion of the sufficiency of the information available to answer the question; and
- o The resolution of the question in terms of either a direct answer or the necessary types of work (e.g. laboratory experiments or field investigations) which need to be conducted to establish the information necessary to satisfactorily answer the question.

Monenco realizes that the standard method of expressing radioactivity in the modern metric system (i.e., the International System of Units) is in becquerels (Bq). Although much of the data presented in this review is in picocuries (pCi), Monenco felt that changing the units to becquerels was outside the scope of the literature survey. The data (and units) which appear in this report are, therefore, exactly as they appear in the literature.

Additional literature which was reviewed for this study but found to be of little use in addressing the specific questions is presented in Appendix A of this report.

PART 2

THE RADIONUCLIDES OF CONCERN

PART 2 - THE RADIONUCLIDES OF CONCERN

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2.1 WHAT RADIONUCLIDES SHOULD BE STUDIED?

211 LITERATURE PERTINENT TO THE QUESTION

Uranium ores always contain the uranium and actinium decay series. In addition, some ores also contain natural thorium (and the thorium decay series). While these decay series contain from 11 to 14 daughter radionuclides each, only a few of these isotopes have half-lives greater than a few months. The long-lived radionuclides, their respective decay series and half-lives are given in Table 2-1.

Radium-226 is the isotope of most concern in the mining and milling of uranium bearing ores and the associated solid and liquid wastes because of its high radiotoxicity and long half-life (1620 years).

If natural thorium is present in the ore, ^{228}Ra may contribute significantly to the effluent radioactivity. Both ^{231}Pa and ^{227}Ac are members of the actinium decay series and have activities of about 1/20 of the uranium decay series members. It is, therefore, unlikely that ^{231}Pa and ^{227}Ac contribute to over-all radioactivity of an effluent.

Uranium-234 is important because it will have an activity about 20 times that of ^{235}U. Adequate control and identification of ^{238}U will also assess the ^{234}U contribution to the over-all radioactivity (Moffett 1977b).

According to Wiles (1983), ^{230}Th and ^{232}Th are of environmental and public health concern because they generate ^{226}Ra and ^{228}Ra. Even though its mass is insignificant, the fate of polonium is very important because in radiation dosimetry of the ^{210}Pb-^{210}Bi-^{210}Po series, polonium's alpha particle contributes more than 95% of the biologically effective radiation dose (Holtzman 1976).

TABLE 2-1

Radionuclides Potentially Associated with Uranium
Mining and Milling Operations and Their Respective Half-Lives*

Radionuclide	Decay Series	Half-life (years)
Radium-226	Uranium	1620
Radium-228	Thorium	5.7
Lead-210	Uranium	22
Uranium-238	Uranium	4.5×10^9
Polonium-210	Uranium	0.4
Protoactinium-231	Actinium	3.4×10^4
Actinium-227	Actinium	22
Thorium-230	Uranium	8.3×10^4
Thorium-232	Thorium	1.4×10^{10}
Uranium-234	Uranium	2.5×10^5
Uranium-235	Actinium	7.1×10^5
Thorium-228	Thorium	1.9

* from Moffett 1977b

The U.S. Nuclear Regulatory Commission identified the key radionuclides to be included in a liquid effluent monitoring program for uranium mills to be natural uranium, ^{230}Th, ^{226}Ra, and ^{210}Pb. Other radionuclides such as protoactinium were not included because the usefulness of the data did not justify the expense of monitoring these radionuclides.

Ryabchikov and Golbraikh (1969) considered five of the six naturally occurring thorium isotopes as quantitatively unimportant. Only ^{232}Th was considered the major isotope of concern.

All six thorium isotopes which occur in uranium and thorium ores contribute to the radioactivity in the tailings. The typical contributions of the six isotopes to the total thorium activity in the tailings are: ^{234}Th (2%), ^{230}Th (77%), ^{231}Th (less than 0.1%), ^{227}Th (4%), ^{232}Th (8.5%) and ^{228}Th (8.5%). These contributions coupled with the maximum permissible concentrations in water of these isotopes identify ^{230}Th, ^{232}Th and ^{228}Th as the thorium isotopes of concern (Moffett 1977a).

Dory (1981) stated that ^{226}Ra was of main concern for the purpose of impact assessment related to aquatic releases from uranium mining and milling.

The principal sources of radiation hazard in the uranium mining and milling industry, according to Dreesen et al. (1982), are those associated with the progeny of ^{238}U. Of particular concern is the release of ^{226}Ra and ^{210}Pb. In addition to these radionuclides Wrenn and Jinks (1976) also considered ^{230}Th to be a potential hazard in terms of human exposure.

212 SUFFICIENCY OF INFORMATION

There is agreement among the experts concerned with the environmental impact of uranium mining and milling that radium (both ^{226}Ra and ^{228}Ra), uranium (^{238}U and ^{234}U), ^{210}Pb and thorium (at least ^{230}Th and possibly ^{232}Th) should be targeted as the radionuclides of concern. There is less agreement on the need to study ^{210}Po. However, since it is always associated with ^{210}Pb, it would seem easy to include it in any over-all study of uranium mining and milling impacts in the aquatic environment. In support of its inclusion, is the fact that its fate in the aquatic environment is known to be associated with organic carbon cycling.

213 RESOLUTION OF THE QUESTION

The radionuclides which should, as a minimum, be studied as part of any impact analysis of the uranium mining and milling on the aquatic environment are:

- ^{226}Ra;
- ^{228}Ra (if ore contains natural thorium);
- ^{238}U and ^{234}U;
- ^{210}Pb;
- ^{228}Th, ^{230}Th and ^{232}Th; and
- ^{210}Po.

It is generally agreed that radium (either ^{226}Ra and/or ^{228}Ra) should receive top priority in the investigation of uranium mining and milling impacts on the aquatic environment. Uranium (either total or as ^{234}U) would rank second in priority. There is less agreement on the assignment of investigation priorities for the remaining radionuclides. From the available literature, ^{210}Pb would appear to be third in line after uranium. Whether or not thorium isotopes or ^{210}Po would be next is

open to conjecture. Polonium has the shortest half-life (0.4 years) and yet contributes 95% of the biologically effective radiation dose of the ^{210}Pb-^{210}Bi-^{210}Po series (Holtzman 1967). Thorium (^{230}Th) has been identified as a key radionuclide of concern in liquid effluents from uranium mills and has a much longer half-life (8.3×10^4 years). However, thorium is notorious for its inability to stay in solution.

PART 3

THE FORM TAKEN BY RADIONUCLIDES ENTERING THE AQUATIC ENVIRONMENT

PART 3 - THE FORM TAKEN BY RADIONUCLIDES ENTERING THE AQUATIC ENVIRONMENT

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3.1 WHAT PROPORTIONS OF THE TYPICALLY OCCURRING DISSOLVED RADIONUCLIDES ENTERING RECEIVING WATERS FROM URANIUM TAILINGS ARE IN THE IONIC FORM AS OPPOSED TO AN ORGANIC COMPLEX?

311 LITERATURE PERTINENT TO THE QUESTION

The form taken by radionuclides entering the aquatic environment from uranium tailings is determined by the mobility of the specific elements within the tailings. Two factors are of paramount importance in determining mobile elements in tailings:

- the mineralogy of the mined uranium ore and deposited tailings; and
- the uranium extraction process (acid or carbonate) (Dreesen et al. 1983).

An investigation into the chemical and mineralogical location of radium and thorium in the uranium mine tailings from Beaverlodge, Saskatchewan was conducted by Wiles (1983). For these particular tailings, radium was located in at least two different physical surrounds:

- one location allows radium to participate in rapid surface exchange reactions with alkali and alkaline earth cations; and
- a second allows release of radium into solution only after a diffusion limited migration.

Thorium was also found in two locations within the tailings material. The first is on the surface of the tailings particles. However, it is not clear whether the initial rapid release of thorium is by surface exchange or by dissolution of a thin surface layer. The

second physical location within the tailings material appears analogous to that of radium. Although a mathematical model was derived for surface exchange and diffusion of radium and thorium from these specific tails, it seems unlikely that a universally applicable quantitative description of the leaching kinetics can be obtained.

The investigation by Moffett and Tellier (1978) of the uranium tailings in the Elliot Lake area demonstrated that pyrite (comprising 4.18% of the tailing) has generated acid within the tailings resulting in the release by leaching of ^{210}Pb, ^{232}Th, ^{230}Th, ^{228}Th, uranium, and ^{226}Ra from the tailings solids. The acidity of the interstitial tailings waters ranged from less than 50 to 15,580 mg/L. Under conditions such as these, the radionuclides of concern would most likely enter aquatic environments in the ionic form of the element. However, no one particular study has confirmed this. It is known that tetravalent uranium (U^{+4}) is found in environments where a reducing electrochemical potential is maintained (Sheppard 1980a). Brown et al. (1981) in a discussion of the mine wastes at Elliott Lake stated that the uranyl complexes (i.e. ionic complexes) are very mobile and readily leached from the strongly acidic oxidizing zone of the tailings.

The movement of radionuclides from tailings to aquatic environments under neutral or basic conditions depends to a great extent on the composition of the water flowing over or through the tailings. Groundwater may be high in minerals (e.g. carbonates) which may mobilize certain radionuclides as complex inorganic ions. Surface water with dissolved complex organics flowing over or through uranium mine tailings has the potential for mobilization of radionuclides by the formation of organometallic complexes.

Under neutral to basic conditions, and lacking significant dissolved organic input, the radionuclides may enter the aquatic environment as complex inorganic ions. Hexavalent uranium is invariably found associated with oxygen as the very stable uranyl ion (UO_2^{+2}) (Sheppard

1980a). These uranyl ions form soluble complexes with naturally occurring anions (e.g. carbonate, sulphate, and carbonate). The uranyl carbonate ions are negatively charged, very stable and tend to predominate in natural waters. The carbonate complexes confer considerable solubility to the uranyl ion over a wide pH range and most uranium is mobilized by this mechanism (Brierley and Brierley 1981). The unusually high concentrations of uranium, thorium and protoactinium in the water of Mono Lake, California have been attributed to the high concentration of carbonate ions in this lake (Simpson et al. 1982, Anderson et al. 1982).

There are various forms of uranium in natural waters depending on the composition and pH of the solution:

- the uranyl-carbonate complex $UO_2(CO_3)_3^{-2}$ in water with elevated levels of bicarbonate and carbonate ions; pH 4.5-6.5;
- the UO_2^{+2} and $UO_2(OH)^+$ ions at a pH between 4.5-7.5 in the form of sulphate compounds in acid solutions (pH less than 4); and
- in the form of organic compounds in weak and acid waters (Ferronsky and Polyakov 1982).

While six isotopes of thorium occur in nature, only the tetravalent form occurs in aqueous solutions. The tetravalent ions have the ability to form many water insoluble compounds. Thorium hydroxide, fluorite, iodate, and phosphate are weakly soluble compounds. Thorium carbonate is poorly soluble in water but is readily dissolved in an excess of alkali carbonate forming a stable complex. In aqueous solutions at pH less than five, thorium is most likely to be present in the ionic and molecular complexed forms. At pH greater than five, the colloidal form of its existence is predominant. The element has a strong tendency towards hydrolysis and absorption and is liable to form complex compounds. The migration of thorium in groundwaters occurs in the colloid-suspended form or in the form of complexes of anionic character,

most probably of organic origin. The exact nature of migrating thorium in surface and groundwaters is not well known (Ferronsky and Polyakov 1982). Because of its ability to be hydrated in natural aqueous systems thorium is easily precipitated or strongly sequestered by surfaces (Koczy 1963; Turekian and Cochran 1978).

Radium is a typical alkaline-earth metal which exhibits only one oxidation state in solution (Ra^{+2}), is highly basic and not easily complexed (Sheppard 1980b). Radium nitrate, chloride, sulphite, and bromite are soluble in water, while the carbonate, sulphate, iodate, and oxalate are insoluble. Usually radium does not form part of the mineral crystalline lattice and thus it can be washed out and transported to natural waters. The radium content of natural waters is usually so small that it never reaches the solubility limits. The unstable state of radium in all natural waters (except where large chloride concentrations exist) leads to its adsorption onto the surface of a solid phase (Ferronsky and Polyakov 1982).

If the water flowing over or through the uranium tailings contains biopolymers (i.e. polysaccharides, polypeptides, or humic materials) which are polyelectrolytic in character, the potential exists for interaction with the radionuclides through ion exchange, chelation, or surface adsorption, resulting in the formation of metal-organic complexes. Uranium-humic complexes have great stability and are related to factors such as steric hinderance, neutralization of charge, and chelation rings (Brierley and Brierley 1981). The very strong binding of uranyl ions to humic acid molecules at acid pH values produces geochemical enrichment factors of about 10,000:1 (sorbed uranyl ion:free ionic uranyl ion) (Szalay 1964). Radium is associated with insoluble organic matter (Sheppard 1980b) and therefore does not generally appear to be associated with dissolved biopolymers in natural water. Thorium, however, does form complexes with organic acids in solution.

The thorium content of ground water was reported to be highest in low salinity, low hardness, low pH, and high organic content groundwaters. These characteristics suggest transport of thorium as colloidal suspensions and anionic complexes involving soil acids (Onishi et al. 1981).

312 SUFFICIENCY OF INFORMATION

Sufficient data does not exist to specifically address the question: What proportions of the typically occurring dissolved radionuclides entering receiving waters from uranium tailings are in the ionic form as opposed to an organic complex?

313 RESOLUTION OF QUESTION

To answer the question in Section 3.1 two types of samples must be taken at uranium tailings sites:

- o groundwater samples from within the tailings; and
- o surface samples of water taken just prior to their entry into the aquatic environment.

These samples should be taken at sites of known tailings mineralogy and uranium extraction process. A minimum of three samples should be taken at each sampling site on the tailings.

Analysis of the water samples must be conducted for:

- o pH;
- o acidity;
- o alkalinity (carbonate, bicarbonate); and

- o total organic carbon, in addition to the radionuclides of concern.

Some method must be found to separate free ionic radionuclides from any organic complexes. These methods may include, but are not limited to, centrifugation, ultrafiltration, filtration through ion exchange membranes, gel filtration or the use of dialysis membranes for the partitioning of inorganics from organic (Benes et al. 1976). The subsequent analysis of the radionuclides in both fractions and in the total sample will then provide the proportions of dissolved radionuclides entering the aquatic environment as an organic complex.

3.2 WHAT VARIATIONS OCCUR IN THE IONIC AND ORGANIC COMPLEX PROPORTIONS?

321 THE LITERATURE PERTINENT TO THE QUESTION

As stated in the previous section (3.1) there is no specific information on the form (i.e. ionic versus organic complex) taken by radionuclides entering natural waters from uranium tailings. Likewise, there is no information on the variation of these proportions.

322 SUFFICIENCY OF INFORMATION

Sufficient data does not exist to specifically address the question.

323 RESOLUTION OF QUESTION

The data required to answer this question must be obtained as outlined in Section 313 (i.e. what form is taken by radionuclides) but on a seasonal basis. More specific information concerning variations in the ionic and organic complex proportions would require more frequent sampling (i.e. bimonthly, monthly, biweekly, weekly).

3.3 WHAT SPECIFIC ORGANIC SPECIES ARE INVOLVED AND WHAT ARE THE MOST LIKELY ORGANIC REACTIONS TAKING PLACE?

331 LITERATURE PERTINENT TO THE QUESTION

The bulk of dissolved organic matter in natural waters consists of highly oxidized, chemically and biologically stable polymeric compounds closely resembling soil humic substances (Reuter and Perdue 1977). These aquatic humic substances are an extremely complex and diverse group of organic materials whose structure is poorly defined. They originate as biodegradation resistant decomposition products and by-products of natural organic matter produced by both plants and animals. Humic substances are arbitrarily divided into three diverse groups of compounds based on their solubility (and insolubility) in dilute acid and base:

- Fulvic acids are soluble in both dilute acid and dilute base.
- Humic acids are soluble in dilute base, but precipitated by dilute acid.
- Humin is insoluble to both dilute acid and dilute base (this fraction is sometimes included with humic acid by some authorities).

The fulvic acid fraction appears to be the predominant group of humic substances in natural waters. Fulvic acids appear to have lower molecular weights than humic acids and contain a higher percentage of oxygen.

Although the exact chemical structure of humic substances (i.e. including all three fractions) is not known, major functional groups have been defined and are reported to be:

- o carboxyl;
- o phenolic;
- o alcohol;
- o methoxyl;
- o carbonyl;
- o ether; and
- o ester.

Unquestionably, humic substances have the ability to react with metals (Snoeyink and Jenkins 1980). However, specific studies concerning the interactions of ^{226}Ra, ^{210}Pb, ^{238}U, or ^{230}Th with humic substances were not located during the literature search. Sheppard et al. (1980) discussed the distribution of ^{234}U and ^{241}Am among soil fractions (including humic acids) and demonstrated that the radionuclide-humate complexes dominate the distribution ratios for the soil systems studied.

The nature of the association between the metal and humic substances is not well understood, however, two modes of binding appear significant:

1) the formation of complexes or chelates between the functional groups of the humic substance and the metal; and

2) an association between the humic substances and a colloidal particle of metal hydroxide (possibly through sorption on the

surface of the particle) whereby the colloidal particles are peptized (i.e. stabilized) in suspension and prevented from coagulating or settling.

The formation of one or another of these humic-metal associations may depend on the initial states of humic matter and metal and their absolute and relative concentrations (Snoeyink and Jenkins 1980).

The data generated by Benes et al. (1976) concerning the possible interactions between humus and thorium in lake water is explained by the possible existence of inorganic colloidal forms of thorium. However, this still does not exclude interaction of humic substances and the ionic form of thorium.

Nash et al. (1981) investigated the redox behaviour, complexing, and adsorption of hexavalent uranium by humic acid and selected clays. The uranyl ion was found to be strongly complexed with humic acid due to the high effective charge of the metal and its affinity for oxygen donor ligands. The mobility of the uranyl ion can, therefore, be governed by the mobility of the humic acid. Unlike other actinides, U^{+6} was not reduced by organic and inorganic environmental reducing agents.

Nash and Choppin (1980) studied the binding of Th^{+4} to three humic and two fulvic acids. Their results indicate the formation of two types of thorium binding sites in the humic polymer containing one or two carboxylate groups. The organometallic complex is highly stable and thorium-humate binding constants were observed to increase with increasing ionization of the humic and fulvic polyelectrolytes.

332 SUFFICIENCY OF INFORMATION

Sufficient data exists to answer question 3.3. However, continued research into dissolved humic substances and these interactions with metals is going on at several academic institutions in North America and Europe.

333 RESOLUTION OF QUESTION

The specific organic species involved in the formation of radionuclide-organic complexes are the dissolved humic substances found in naturally occurring waters.

The most likely organic reactions taking place with the radionuclides are:

1) the formation of complexes or chelates between the functional groups of the humic substance and the radionuclide; and/or

2) an association between the humic substance and a radionuclide hydroxide colloid, whereby the colloidal radionuclide particles are stabilized in suspension.

PART 4

THE REMOVAL OF RADIONUCLIDES FROM SURFACE WATERS

PART 4 - THE REMOVAL OF RADIONUCLIDES FROM SURFACE WATERS

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4.1 WHAT ARE THE MECHANISMS OF REMOVAL OF RADIONUCLIDES FROM SURFACE WATERS?

411 LITERATURE PERTINENT TO THE QUESTION

The general mechanisms of removal of radionuclides from surface waters are given by Whicker and Schultz (1982) and are explained in Section 413. More specific information on these mechanisms are given by Turekian (1977), Li *et al*. (1981), Santschi *et al*. (1979), Dyck *et al*. (1971), Sebesta *et al*. (1981) and Sheppard (1980a,b).

412 SUFFICIENCY OF INFORMATION

There is sufficient data to answer the question. The literature relating to the various mechanisms will be reviewed in Sections 4.2, 4.3 and 4.4.

413 RESOLUTION OF THE QUESTION

The removal of metals (including radionuclides) from surface waters involves the following mechanisms:

- chemical adsorption to organic and inorganic particulate material;
- absorption by living particulate material;
- incorporation into zooplankton fecal pellets;
- precipitation scavenging; and

- ion exchange (Whicker and Schultz 1982)

Elaboration on each of these points will be made in Sections 4.2, 4.3 and 4.4.

4.2 WHAT ROLES DO ABSORPTION, ADSORPTION AND PRECIPITATION PLAY IN THE REMOVAL OF RADIONUCLIDES FROM SURFACE WATER?

421 LITERATURE PERTINENT TO THE QUESTION

An analysis of the ^{210}Pb regime in the surface layers of the ocean by Turekian (1977) demonstrated that particles (inorganic) are more important scavengers of many trace metals than the bodies of micro-organisms. The sequestering of trace metals (including radionuclides) by inorganic particulate material and their removal by sedimentation was decreased from a time frame of 12 to 18 months, in the open water of the ocean, to days as coastal areas were approached. This was chiefly due to the greater concentration of inorganic particulate material closer to shore. This same phenonenon should also operate in large oligotrophic lakes.

Li *et al*. (1981) and Santschi *et al*. (1979) demonstrated that removal of ^{238}Th from surface waters was mainly through adsorption onto non-living particles and subsequent settling to the bottom. Removal of ^{228}Th by the phytoplankton-zooplankton-fecal pellet route was relatively small. By contrast, removal rates of ^{210}Po by the phytoplankton-zoo-plankton-fecal pellet route accounted for almost all of the radionuclide removed from surface waters. This removal was coupled with the organic carbon cycle and thus exhibited seasonal variation.

Uranium in surface waters is extremely mobile (especially as a carbonate complex). Under reducing conditions (as might occur in the hypolimnion of a lake) the uranium precipitates as a hydroxide. Uranium hydroxides also tend to precipitate with ferric and manganese hydroxides especially when the pH and carbonate ion concentration drop. Suspended organic matter also provides a surface for this precipitation. Radium-226 is immobile in surface waters because it and the parent thorium form insoluble compounds in natural environs (Dyck et al. 1971).

Uranium retention by suspended sediments in oxidizing and alkaline conditions is poor because the predominant uranium species at pH 6 in oxidizing environments are either neutral or negatively charged. However, in the neutral to acidic pH range, the cation exchange properties of sediments could contribute to the adsorption of uranium on clay minerals due to the presence of the UO_2^{+2}. Adsorption on organic materials and humic substances is, however, very strong, especially at acid pH values (Onishi et al. 1981).

Sebesta et al. (1981) found a significant part of the radium in water pumped from a uranium mine was present in particulate form. Surface adsorption is apparently important for ^{226}Ra in soils. Radium-226 concentrations increased with particle size below 100 um (Sheppard 1980b).

Ion exchange appeared to be the controlling factor in the removal of radium from sea water by deep sea clays (Holland and Kulp 1954). Titaeva (1967) also found that removal of radium by clay sediments and insoluble humic material was due to ion exchange. If high concentrations of calcium carbonate were present ion exchange was reduced and suggested coprecipitation of radium into $CaCO_3$. After reviewing the literature concerning the adsorption and desorption mechanisms for radium Onishi et al. (1981) concluded that radium would be expected to substitute for other divalent cations during replacement or precipitation

reactions. A direct correlation of cation exchange capacity with adsorption and leaching studies with different types of competing cations suggested that an important reaction mechanism of radium adsorption is cation exchange.

In a study of radionuclide speciation in sediments and soils, ^{226}Ra was found to be more readily available for exchange and more likely to be associated with carbonate minerals than ^{210}Pb. The iron content of the sediment appears to influence the manner by which ^{226}Ra and ^{210}Pb are bound to the sediment. Where the iron content is high and in the oxide form, both ^{226}Ra and ^{210}Pb are likely to be adsorbed to this phase (Copper et al. 1981).

The rapid adsorption of ^{210}Pb (from atmospheric ^{222}Rn) onto settling particulates in aqueous environments and their incorporation into accumulating sediments is the basis for a geological clock for establishing the chronology of recent sediments (Durham and Joshi 1981; Copper et al. 1981).

Laboratory studies designed to investigate the bioaccumulation potential of thorium and uranium in rainbow trout (Salmo gairdneri) found that soluble thorium introduced into natural river water rapidly partitioned into soluble and particulate fractions. Thorium had to be continually introduced into the aquaria over the 27 day experimental period to maintain the radionuclide in solution. The adsorption of thorium onto surfaces of the aquarium (as thorium dioxide), uneaten food, algae and fecal material was given as the reason for the actual concentrations never reaching the theoretical levels of exposure (Poston 1982).

Soluble uranium introduced into the natural river water also decreased in concentration over the experimental period and the partial replenishment method also had to be used to maintain dissolved uranium levels. The loss of uranium from solution was explained by precipitation or plating onto aquarium walls (Poston 1982).

In a study of aquatic foodchain uptake and translocation Mahon (1982) found that the algal component of the food chain exhibited the highest concentration of uranium, ^{210}Pb and ^{210}Po. This was apparently due to the surface adsorption of these radionuclides rather than the internal accumulation (i.e. absorption). Radium-226 was found to accumulate in the sediments to a greater extent than any other component, apparently due to adsorption.

Hesslein and Slavicek (1984) demonstrated a rapid removal of ^{226}Ra from surface water to sediments in four lakes of the Experimental Lakes Area (Ontario, Canada). This rapid transfer to the sediments was due to manganese oxides which have a very high affinity for radium. There is a possibility that coprecipitation or adsorption of radium to precipitated ferrous sulphide also serve to transfer radium to the sediments.

In a comparison of the relative mobility of thorium, lead and polonium nuclides (i.e. ^{210}Pb, ^{234}Th, ^{228}Th) in large (13 m^3) marine microcosms and Narragansett Bay (Rhode Island, U.S.A.) all curves for the removal of these radionuclides from the water columns showed an initial decrease in concentration during the first few days (or weeks, depending on season) followed by a period of slower removal (Santschi et al. 1983). The authors hypothesize that removal of the radionuclides from the water column was probably controlled by:

- adsorption of nuclides onto suspended particles followed by gravity settling or filtering by benthic fauna; and

- adsorption on resuspended settled particles.

Removal rates of the thorium isotopes in the natural bay and the microcosms were very similar (one-half removal times varied between one and 10 days and were faster in the summer). However, removal rates for ^{210}Pb were much slower in the bay than in the microcosm. Large

fractions of the radionuclides in the water columns of the microcosms were associated with filterable particles (i.e. greater than 0.4 um), except for ^{228}Th in the summer.

The quantitative evaluation of radionuclides in a water column (as dpm/cm^3) at time t was expressed as the mathematical expression:

$$\frac{dC}{dH} = -\frac{S}{H} C_p + \frac{S}{H} C_m^0$$

where:

- o C = the total concentration (dissolved and suspended) of radionuclide in the water column (dpm/cm^3) at time t
- o H = height of water column
- o S = flux of suspended particles to sediments ($g/cm^2/day$) which also equals the resuspension rate of sediment particles
- o C_p = the concentration of radionuclide in suspended particles (dpm/g) at time t
- o C_m^0 = the concentration of radionuclide in sediment (dpm/g) at depth zero and time t

The rapid removal of ^{228}Th from surface waters of the New York Bight is thought to be through adsorption of the radionuclide onto non-living particles and their subsequent settling to the bottom. These non-living suspended particles are continuously provided by the resuspension of bottom sediments near shore and from river inputs (Li et al. 1981).

Holland and Kulp (1954) studied the uptake of thorium by deep sea clays and concluded that base exchange is the method by which thorium is taken up by sediments. Thorium incorporation into sediments by precipitation mechanisms has also been documented. Laboratory studies also show that thorium tends to precipitate as thorium hydroxide and hydrated thorium oxide even in moderately acidic environments (Onishi et al. 1981).

Additional literature concerning the role of biological organisms in the removal of radionuclides from surface waters (due to absorption or adsorption) will be addressed in Section 4.3

422 SUFFICIENCY OF INFORMATION

Enough information is available in the scientific literature to generally address the removal of radionuclides from surface water by the mechanisms of chemical adsorption, and precipitation scavenging. To a large degree, the mechanisms which are responsible for removal of radionuclides in freshwater must be inferred from marine studies. Specific laboratory studies of radionuclide removal from freshwater by organic detritus appear to be lacking.

423 RESOLUTION OF THE QUESTION

Specific laboratory or field studies should be conducted to quantify the removal of uranium, radium, lead, and polonium from surface fresh waters. Thorium is well known for its inability to stay in solution and does not need to be addressed in these studies. These studies should examine the effects of suspended sediment particle size, organic content, ion exchange potential, water pH, and redox potential on the removal of radionuclides from water by sedimentation.

4.3 WHAT ROLES DO BIOLOGICAL ORGANISMS PLAY IN THE REMOVAL OF RADIONUCLIDES FROM SURFACE WATER?

431 LITERATURE PERTINENT TO THE QUESTION

Biological organisms can remove radionuclides from surface waters by several mechanisms:

- o adsorption to the exoskeleton or cell walls with subsequent sedimentation at death;
- o absorption into the organism with transfer to the sediment at the organism's death as part of the organic detritus;
- o consumption of contaminated detritus or living cells and release in fecal pellets with subsequent sedimentation; and
- o absorption through the gut and sequestering in the organism body.

Adsorption to the Exoskeleton or Cells Walls or Absorption Into the Organism

Strandberg and Shumate (1982) investigated the use of microbial cells for removing uranium, radium and cesium from nuclear processing waste streams. They found that uranium accumulated as needle-like fibrils in a layer approximately 0.2 um thick on the surface of Saccharomyces cerevisiae. Little or no uranium was found within these cells. By contrast, uranium formed dense intracellular deposits in Pseudomonas aeruginosa. The surface-associated accumulation of uranium exhibited by S. cerevisiae is consistent with the view that metal biosorption occurs by the complexation of positively charged metal ions with negatively charged reactive sites on the cell surface or in

extracellular polymers. The uptake of uranium by S. cerevisiae did not require cellular metabolism. The rapid intracellular uptake of uranium by P. aeruginosa suggested involvement of metabolically mediated active transport, however, this was later discounted.

Havlik (1971) and Havlik and Robertson (1971) studied the role of unicellular algae in the uptake of ^{226}Ra from aqueous media. Two types of response to radium addition were demonstrated:

- absortion of radium within the algal cells (the amount absorbed was proportional to the length of exposure); and
- adsorption of radium onto the cell surface or gelatinous capsule.

Although both responses occurred in all cells, absorption predominated in only two of the five species studied, whereas adsorption was more typical of the remaining species. The highest accumulation of radium was reached after 24 hours of exposure, however, the amount of radium accumulated was inversely proportional to the radium concentration of the medium. The mechanisms of radium uptake by algae are affected by:

- the ability of radium to bind to organic matter;
- the ability of radium to adsorb to cell membranes and cell surfaces reversibly; and
- the ability of some algae to absorb and metabolize radium.

No rates of adsorption were given by the authors.

In the studies reviewed, unicellular and filamentous algae exhibited the highest concentrations and bioaccumulations of the

radionuclides ^{210}Pb, ^{210}Po and natural uranium, relative to other aquatic organisms.

Tanaka et al. (1983) observed decreases in ^{210}Po, ^{210}Pb and ^{234}Th in the water column of a marine bay caused by the increased production of organisms during phytoplankton blooms. Subsequent sinking of the organisms associated with the bloom increased the radionuclide content of the bottom water in the bay.

Tsivoglou et al. (1958, 1960) discussed the pollution of streams and rivers by the uranium ore refining industry in the "Four Corners" area of the Colorado River Basin in the western U.S. Algal samples (i.e. periphyton) collected downstream from one mill had 1500 times the radium content of algae collected upstream of the mill. Benthic stream insects and fish also indicated the same general increase of radium downstream from the mill although to a much smaller degree than the algae. As with other studies, the exact mechanism of uptake (i.e. adsorption or absorbtion) was not discussed.

Havlik (1967 and 1970) also described the accumulation of radium in algae, bryophyta and spermatophyta in areas known to have increased concentrations of radium in the environment. Generally, higher accumulations were found for planktonic algae than for aquatic vascular plants. The cause of these increased concentrations (i.e. adsorption or absorption) was not discussed.

Adsorption of several radionuclides onto the chitinous moults of pelagic crustaceans allows the accumulation of trace elements to concentrations in excess of that in the surrounding water. The sinking of these moults provides a mechanism by which they are transported to the sediment (Fowler 1977, Martin 1970).

In a study of the uptake of artificially introduced radium in a small lake, Emerson and Hesslein (1973) found that the algal material

growing on rocks in the littoral zone removed the isotope from the water by adsorption rather than absorption. The differentiation between adsorption or absorption of soluble uranium ions was not made by Brierley et al. (1980) in their discussion of the removal of uranium from solution by filamentous and benthic algae (i.e., Spirogyra and Chara). Marchyulenene et al. (1978) and Marchyulenene (1978) attributed the uptake of ^{210}Pb by the macroalga Nitellopsis obtusa to be caused by the adsorption of the radionuclide on the cell walls. The exact accumulation rate of ^{210}Pb was determined by its physico-chemical state of the water and the chemical properties of the cell walls.

Justyn and Stanek (1974) investigated the accumulation of natural radionuclides by bottom sediments and aquatic organisms in streams both affected and unaffected by uranium ore mining and milling. Their results demonstrated a higher cumulative capacity in algal plankton, periphyton, aquatic bryophytes and macrophytes for ^{226}Ra and uranium than in the upper two centimetres of bottom sediments.

Dusauskene-Duz and Polikarpov (1978) observed that algae accumulate ^{210}Pb more actively and in larger amounts than do macrophytes. Accumulation coefficients (concentration in plant cells/concentration in water) for this radionuclide in Chara contraria were found to be on the order of 1500.

Mahon and Mathewes (1983) found that the accumulation of uranium, ^{226}Ra, ^{210}Pb and ^{210}Po by the yellow pond lily (Nuphar lutea) was due to two types of mechanisms. The rapid accumulation of uranium and radium in the spring (and its subsequent decline in the summer) was a reflection of the active metabolic state of the plant. Thus, absorption and possible metabolism of these radionuclides seems plausible. However such seasonal trend was observed for lead or polonium. The accumulation of the latter two radionuclides was, apparently, due to adsorption to the cell walls of the plant.

The concept of discrimination for or against radium in biologic pathways requiring calcium was discussed by Hesslein and Slavicek (1984). Under this theory, the biological uptake of radium is linked to calcium metabolism wherein individual organisms or processes with organisms have the ability to favour or discriminate against the selection of radium atoms compared with calcium atoms. It appears from the many studies conducted with radium and macrophytes that radium uptake and incorporation is favored over calcium in macrophytes. The same is apparently true for crayfish. However, there is a strong discrimination against radium in fish. This linkage to calcium metabolism is reinforced by earlier observations of the influence of calcium on radium uptake in aquatic plants (i.e. increased calcium concentrations caused a decrease in radium uptake).

In his study of marine plankton radium content Szabo (1967) defined a discrimination factor for radium with respect to calcium as:

$$DF^{m}_{Ca} = A_m / A_{Ca}$$

where:

A_m = accumulation or concentration of radium in the organisms over that in sea water; and

A_{Ca} = accumulation or concentration of calcium in the organisms over that in sea water.

Using discrimination factors, radium was found to be preferred over barium, strontium and calcium.

A strong discrimination for ^{210}Po was observed by Heyraud and Cherry (1979) in the hepatopancreas of marine crustaceans and cephalopods. The levels encountered (3300 to 61000 pCi/kg) were termed

"exceptionally high by standards of the natural radiation environment." This selective accumulation of ^{210}Po was dependent on the concentration of ^{210}Po ingested in the days immediately prior to radionuclide determination.

Removal by Fecal Pellet Formation

Whether absorbed intracellularly or adsorbed to the outer cell wall, radionuclides can be transported by phytoplankton to the sediment directly by the death of the cell or by its incorporation into fecal pellets after it is consumed by zooplankton. Studies concerning marine zooplankton fecal pellets have shown that they sink fairly rapidly (50 to 950 metres per day) and the membrane surrounding the material decomposes slowly. Fecal pellets have been shown to be important in the vertical oceanic transport of polonium (Cherry et al. 1978).

Li et al.'s (1981) study of radionuclides in the waters of the New York bight demonstrated that the removal of ^{210}Po from the surface waters was almost entirely associated with the phytoplankton-zooplankton-fecal pellet pathway. This same mechanism may be responsible for the removal of ^{210}Po from the limnetic zone of lakes.

Smayda (1969) determined the sinking rates of fecal pellets collected from seawater and compared them with rates for sinking fish eggs, phytoplankton, protozoans and zooplankton. Sinking rates for fecal pellets ranged from 36-376 m/day and generally were faster than literature values for living and dead phytoplankters. However, Smayda (1969) could not determine if fecal pellets represented a significant transport mechanism for organic material to deep water. An overview of the role of zooplankton fecal pellets in aquatic ecosystems is given by Turner and Ferrante (1979).

Beasley et al. (1978) determined the activities of ^{210}Po and ^{210}Pb in fecal pellets from the marine zooplankter Meganyctiphanes

norvegica. The ^{210}Po:^{210}Pb activity ratio in the fecal pellets was in good agreement with that value obtained from suspended particulate matter in surface seawater. Estimates of ^{210}Po and ^{210}Pb removal times from the surface mixed layer by fecal pellets alone yielded values which were of the same order of magnitude as the removal times for these radionuclides by all routes. It was suggested by the authors that zooplankton fecal pellets play a significant role in the removal of ^{210}Po and ^{210}Pb from the surface layers of the ocean.

Absorption Through the Gut and Sequestering in the Organism Body

An investigation into the bioaccumulation potential of thorium and uranium in rainbow trout (Salmo gairdneri) by Poston (1982) concluded that the primary input of uranium and thorium into the test fish was by gut absorption (as opposed to gill absorption). Even in this case however, the actual tissue-bound fraction was relatively small and the majority of the total body burden was located in the gastro-intestinal tract. The small fraction which becomes fixed in the carcass is most likely located in the bone.

Fish inhabiting a river contaminated by uranium tailings contained less ^{226}Ra per gram of ash (as whole fish samples) than algae and insects inhabiting the same river. There was also a considerable difference in radium concentration among the fish species sampled. Speckled dace (Rhinichthys osculus) contained more radium than other fish in the tailings impacted area (as much as 16 times the radium concentration in the unimpacted upstream area). Within the impacted area, dace (which eat algae) had higher concentrations of radium than the sculpin (Cottus sp.) which consumed mainly insects. Funnelmouth suckers (Catostoma latippinis) also contained radium in concentrations similar to the dace. This was a reflection of the feeding habit and ingestion of significant amounts of sediment by this species (Sigler et al. 1966). Not enough large fish were collected during the study to generate reliable data concerning the concentrations of radium in the flesh and

skeleton. Organs and viscera were discarded without radionuclide analysis.

Emery et al. (1981) studied the uptake of ^{238}U from a contaminated nuclear waste pond in the northwestern U.S. Direct sediment-to-fish transfer was concluded to be the primary route for uranium uptake from the pond environment. Most of the contamination in the exposed fish was located in the gut or on external surfaces.

Three fish species inhabiting a lake in northern Saskatchewan which received untreated uranium mill tailings had significantly higher concentrations of ^{226}Ra, ^{210}Pb and uranium than fish from a control lake (uninfluenced by uranium mill tailings). Accumulation differed among fish species with bottom-feeding white suckers (Catostomus commersoni) accumulating the highest concentrations. This was probably due to their close association with the radionuclide contaminated sediments. Omnivorous lake whitefish (Coregonus clupeaformis) had the second highest radionuclide concentration. The inclusion of phytoplankton and zooplankton (known to concentrate radionuclides from the water column) in their diet probably assisted in the bioconcentration of radionuclides in this fish species. By contrast, the piscivorous lake trout (Salvelinus namaycush) exhibited the least accumulation exemplifying the lack of transfer of these radionuclides through the various trophic levels in the food chain (Lush and Swanson 1981). Although the authors state that the accumulation of radionuclides was highest in the skin followed closely by bone, no indication was given as to the concentration of radionuclides in the gut which should have been extremely high for the suckers.

In an examination of surface streams affected by uranium mining and milling (Justyn and Lusk 1976) a comparatively smaller portion of radium and uranium was deposited in the musculature compared with the skeleton and entrails.

432 SUFFICIENCY OF INFORMATION

Enough information is available in the scientific literature to address the question concerning the role of biological organisms in the removal of radionuclides from surface waters.

433 RESOLUTION OF THE QUESTION

Biological organisms can remove radionuclides from surface waters by several mechanisms:

- o adsorption to the exoskeleton or cell walls and their subsequent sedimentation at death;
- o absorption into the organism with transfer to the sediment at the organism's death as part of the organic detritus;
- o consumption of contaminated detritus or living cells and release in a fecal pellet with subsequent sedimentation; and
- o absorption through the gut and sequestering in the organism body.

More specific information concerning these mechanisms is given in Section 431.

4.4 WHAT IS THE RELATIONSHIP BETWEEN THE PELAGIC, PROFUNDAL AND LITTORAL ZONE OF A LAKE IN THE REMOVAL OF RADIONUCLIDES FROM SURFACE WATER BY BIOLOGICAL ORGANISMS?

441 LITERATURE PERTINENT TO THE QUESTION

The term pelagic is widely used in ecology to describe the zone of the sea or ocean which includes all life in the open water (i.e. plankton, nekton and neuston) (Odum 1971). A comparable limnological term would be the limnetic zone defined as the open-water zone to the depth of effective light penetration. The profundal zone is the bottom and deep water area beyond the effective light penetration. The littoral zone, typically occupied by rooted plants, is the shallow-water region with light penetration to the bottom.

The biological removal of radionuclides from each of these zones depends on the number and type of organisms inhabiting these zones.

Biological Removal From the Limnetic Zone

The limnetic zone is occupied by phytoplankton, zooplankton, and fish. Studies in the scientific literature have demonstrated that radionuclides are both adsorbed and absorbed by planktonic algae. Phytoplankton and unicellular algae in general seem to accumulate more radionuclides per unit weight than any other aquatic organism. Death and sinking of these algae removes the radionuclides from the water.

In addition, zooplankton are known to remove radionuclides by two mechanisms:

- ingestion of algae containing radionuclides and subsequent incorporation of gut contents into a fecal pellet and removal through sinking; and

- adsorption of radionuclides onto the exoskeleton with removal after moulting and sinking.

No studies were found in the literature survey which reported on the amount of radionuclides absorbed by zooplankton feeding on radionuclide contaminated food.

From the available scientific literature, the main loading of radionuclides to fish is via the gastrointestinal tract. Those radionuclides not absorbed by the gut lining would probably be adsorbed to the organic matter in the gut and pass out of the fish with the fecal material.

Biological Removal From the Profundal Zone

In comparison to the limnetic zone, very little biological activity occurs in the profundal zone. Some blue-green algae (or bacteria) are known to exist for at least part of a 24-hour cycle below the depth of effective light penetration in some lakes. Zooplankton may inhabit the upper portion of the profundal zone during the day and some types of fish may also be present for at least part of the 24 hour cycle. The mechanisms by which these organisms would remove radionuclides from the water would be the same for the limnetic zone, albeit on a reduced scale.

Biological Removal From the Littoral Zone

Compared to both the limnetic and profundal zones, the littoral zone has the potential to contain a much wider variety of organisms. These organisms may be divided into the following groups:

- rooted and floating aquatic vascular plants;

- macroalgae (e.g. Characeae or Cladophorales) attached to the littoral substrate;
- microalgae attached to rock and other nonliving substrates;
- microalgae attached to living macrophytes (i.e. aquatic vascular plants and macroalgae);
- littoral phytoplankton;
- littoral zooplankton;
- littoral immature insects (e.g. dragonfly larvae, blood worm larvae, mayfly larvae, etc.);
- littoral adult insects (e.g. herbivorous and predaceous water beetles, hemiptera such as water boatmen and water striders);
- littoral mollusca (e.g. clams and snails);
- other littoral invertebrates (e.g. aquatic nematodes, oligochaetes, aquatic mites); and
- small fish.

Thus, the variety of living organic surfaces available for adsorption or absorption in a well developed littoral zone is much greater than either the limnetic or profundal zone. Although all components of the littoral zone have not been investigated for their ability to remove radionuclides, aquatic vascular plants, macroalgae, and attached microalgae have been demonstrated to concentrate at least some of the radionuclides of concern in this literature review (especially ^{226}Ra) and provide an important pathway for their removal from the water. The individual organisms were discussed in Section 4.3

In summary, the littoral zone of a lake provides the potential for a much greater removal of introduced radionuclides from the surface waters. Once these littoral organisms die and become detritus, there is a general tendency for this material to be shifted toward the centre or deeper portions of the lake in a phenomenon known as sediment focusing. From the available literature, it seems that the radionuclides bound to this organic detritus (either through physical adsorption or chemical binding) remain bound and follow the detritus into sediment incorporation.

This discussion is, of course, contingent upon the fact that the littoral zone is well developed. Some lakes will have wave impacted littoral zones wherein conditions are physically too harsh for the maintenance of rooted vascular plants or attached macroalgae. Pure sand substrates in the littoral zone also prevent the colonization by significant amounts of rooted plants or attached algae. In cases such as these, the littoral zone will become much less important in the removal of radionuclides compared to the limnetic zone.

442 SUFFICIENCY OF INFORMATION

Although all components of a well developed littoral zone have not been investigated to assess their role in the removal of radionuclides from water, enough information is available to address the question: What is the relationship between the pelagic, profundal and littoral zone of a lake in the removal of radionuclides from surface water by biological organisms?

443 RESOLUTION OF THE QUESTION

The littoral zone of a lake has the potential to remove larger quantities of radionuclides from surface waters than either the limnetic

or profundal regions of the same lake). The limnetic and profundal zones do not exhibit the wide variety of biological organisms normally associated with the littoral zone. Of major importance in the removal of radionuclides from surface waters are attached algae and rooted vascular aquatic plants, both of which form a major portion of the inhabitants in a well developed littoral zone.

PART 5

SEDIMENTATION OF RADIONUCLIDES AND DISPERSION WITHIN SEDIMENTS

PART 5 - SEDIMENTATION OF RADIONUCLIDES AND DISPERSION WITHIN SEDIMENTS

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5.1 WHAT IS THE SEDIMENTARY HISTORY FOR AREAS KNOWN TO BE AFFECTED BY URANIUM MILLINGS OPERATIONS?

511 LITERATURE PERTINENT TO THE QUESTION

The work of Zaidi (1980) and Neame et al. (1982) are the only sediment core sampling programs known to have been carried out in a lake, which received waters from uranium milling operations. Both studies were for the purpose of determining radionuclide concentrations within successive layers of sediment cores. The results obtained by Zaidi (1980) were questionable due to thawing of the frozen sediment cores and mixing of the material in transit to the laboratory (Saskmont Engineering Limited 1980). Neame et al. (1982) examined the concentrations of ^{210}Pa, ^{210}Pb, ^{228}Th, ^{230}Th, and ^{226}Ra in successive layers of frozen sediment cores from areas impacted by uranium mining. Generally, the four cores, 10 cm in depth, taken in a bay receiving the discharge from the mill tailings treatment area had increased concentrations of ^{210}Pb and uranium in the top two centimeters section. The middle section of the cores from this bay contained slightly elevated concentrations of ^{226}Ra. It is not known whether these concentrations are significantly higher than the concentrations at the bottom of the core. The 18 cm deep cores taken in a lake receiving runoff from waste rock disposal areas contained increasing concentrations of ^{228}Th, ^{236}Th, ^{226}Ra, ^{210}Pb and uranium in the two centimeter sections from the bottom of the core to the top. However, no time-depth correlation which could be used for sedimentary history was developed for these cores.

512 SUFFICIENCY OF INFORMATION

A review of published and unpublished data demonstrate that existing data are not sufficient to answer the question (5.1). There has

apparently never been a detailed study of the sediment cores from any lake impacted by uranium mining and milling relating to the sedimentary history of the lake.

513 RESOLUTION OF THE QUESTION

The study of the sedimentary history of areas known to be affected by uranium mining and milling operations will involve the collection of a number of core samples from various parts of the lake basin. From these cores, qualitative stratigraphic changes must be identified and correlated with known historical dates. More details relative to the resolution of the question will be given in the following sections.

5.2 CAN SEDIMENT CORES COLLECTED FROM THOSE AREAS BE USED TO DEFINE A SEDIMENTARY HISTORY THAT WILL BE USEFUL IN PREDICTIVE MODELLING?

521 LITERATURE PERTINENT TO THE QUESTION

No information was found in the literature review which could be used to address this question.

522 SUFFICIENCY OF INFORMATION

Insufficient information is available to address this question.

523 RESOLUTION OF THE QUESTION

The only way to determine if sediment cores from areas affected by uranium mining and milling operations can define a sedimentary history for use in predictive modelling is to take cores in these areas. Even if a time-depth relationship can be established, deposition rates measured directly from a core are subject to variations of basin shape and sediment deposition. Currents may strongly influence disposition of lake sediment. In addition, sediment focusing may cause more sediment to be deposited in the deeper parts of the basin. Sediment focusing may also change over time (Lehman 1975, Davis and Ford 1982). Because of the variety of phenomena which can effect sediment deposition, lakes influenced by uranium mining and milling operations must have multiple sediment cores taken throughout the basin and analyzed for time-depth relations. Mathematical models are available for the determination of sediment accumulation from single core measurements and their transformation to basin-wide influx rates. Distortions due to sediment focusing can also be assessed (Lehman 1975).

5.3 WHAT TECHNIQUES SHOULD BE USED TO OBTAIN SAMPLES FOR HISTORICAL ANALYSIS?

531 LITERATURE PERTINENT TO THE QUESTION

Perhaps the best example of sedimentary history reconstruction is that of Davis and Ford (1982) concerning Mirror Lake, New Hampshire, U.S.A. Twenty-three cores were taken at various depths from different parts of the basin in the 15 ha lake. Cores were collected with a Livingstone piston corer, a Davis peat sampler (both described in Wright et al. 1965) as well as a Swedish foil corer. Probings of the lake sediment was also accomplished at an additional 27 sites.

The age of the sediment at many levels (for selected cores) was established by radiocarbon dating, or by cross-dating pollen analyzed cores to pollen analyses from radiocarbon dated cores. Rates of sediment accumulation (as $mg/cm^2/yr$) could be compared throughout the lake's history in different parts of the basin (Davis and Ford 1982).

In areas influenced by uranium mining, calculation of sedimentation rates based on ^{210}Pb would be useless due to the influx of the radionuclide from the mining operation. Non-destructive techniques such as X-ray scanning, densitometry, whole core-magnetic susceptibility measurement and isothermal remnant magnetization may be used to define the various layers of sediment, but some method (i.e. carbon dating, palynology, or ^{137}Cs dating) would still have to be used to establish a time-depth relationship for each core. Once this was established, the depth of some radionuclides within the core (i.e. those not diffusing within the core) could be related to the known start-up date of the mining operation. An important aspect of any study to identify the sedimentary history of areas influenced by uranium mining is the study of control areas in the same region. Background radionuclides in these "uncontaminated" sediments may play an important role in defining the sedimentary history of the areas known to be influenced by mining.

Although some concern about collecting undisturbed sediment cores has been expressed by Baxter et al. (1981) the use of the Craib corer for obtaining undisturbed sediment cores has not found wide acceptance.

532 SUFFICIENCY OF INFORMATION

There exists a sufficient amount of data concerning the collection of sediment cores and their stratigraphic analysis for use in the collection and historical analysis of sediments core samples from lakes impacted by uranium mining and milling. However, the operation of

uranium mines is a very recent chronological event in the history of some lakes and natural sedimentation rates may not be sufficient to delineate this occurrence.

533 RESOLUTION OF QUESTION

Although a variety of coring devices exist, the Livingstone piston corer seems to have obtained favour with a number of paleolimnologists and has been used to collected sediment cores of four to six metres in length (Hickman and Klarer 1981; Forbes and Hickman 1981). Prior to coring, a bathymetric survey of the lake should be conducted and areas of accumulated sediments identified. Probing of sediment depths at various locations in the lake is recommended. In addition to the deepest portion of the lake, additional cores should also be taken at various depths in different portions of the lake basin. It may not be necessary to analyze all cores taken, but their visual inspection and availability for analysis, if needed, will enhance the results of sediment core analysis.

Natural radionuclide deposition cannot be used as a tool for obtaining time-depth relationships. Other proven methods which should be considered for use are ^{137}Cs dating, radiocarbon dating or palynology. It may be that a combination of all three methods is required to obtain a time-depth relationship.

5.4 WHAT SPECIFIC LOCATIONS COULD BE USED TO EFFICIENTLY GAIN AN UNDERSTANDING OF PAST SEDIMENTATION RATES?

541 LITERATURE PERTINENT TO THE QUESTION

Many investigators believe that variations in core stratigraphy due to differential inputs of sediment material around the lake can be avoided by coring near the deep or central portion of the lake. However many exceptions to this general belief are known (Davis and Ford 1982). The Mirror Lake study involved the collection of one central core at the deepest portion (i.e. 10 m) of the 15 ha lake and 22 additional cores from depths ranging from 2 to 9 metres. Sample cores were taken along both major axes of the lake as well as in prominent shallow areas. Data from these shallow cores, the main, central core and four cores of intermediate depth (i.e. 6.2, 6.8, 7.2 and 8.9 m) were studied in detail. The results of this study demonstrate that the past sedimentary history of a lake determined from a single sediment core may be misleading. Settling rates of different types of sedimentary material may follow different patterns within the lake basin. Sediment focusing may have also changed through time reflecting a change in the water circulation and volume of water entering the lake. In Mirror Lake, the shifting pattern of sediment focusing made it impossible to interpret the fluxes of sediment to the basin from a single core.

542 SUFFICIENCY OF INFORMATION

To date, too few detailed studies have been completed to specifically locate the sites to be used for sediment coring in a lake without previous core sampling.

543 RESOLUTION OF QUESTION

Detailed bathymetric and sediment core studies must be conducted on each individual lake before it will be possible to define specific locations (other than the deepest point) for an understanding of past sedimentation rates.

5.5 WHAT ARE THE CURRENTLY EXISTING SEDIMENTATION RATES OF RADIONUCLIDES?

551 LITERATURE PERTINENT TO THE QUESTION

Since the initial use of ^{210}Pb for sediment geochronolgies in both the marine and freshwater environments there has been a great deal of attention focused on measuring the flux of this particular radionuclide into the sediment. The review by Robbins (1978) summarizes the flux of ^{210}Pb (in $dpm/cm^2/yr$) to lake sediments along with average sedimentation rates (in mm/yr and/or $mg/cm^2/yr$) in 23 published studies. Lead-210 flux ranged from 0.42 to 2.29 $dpm/cm^2/yr$ and average sedimentation rates ranged from 0.1 to 40 mm/yr.

Durham and Joshi (1981) demonstrated the excess flux of ^{210}Pb (that amount of ^{210}Pb not supported by ^{226}Ra) to the sediments of Lake Superior ranged from 0.17 to 0.41 $pCi/cm^2/yr$ with corresponding sedimentation rates of 16.7 and 29.5 $mg/cm^2/yr$.

Robbins and Edgington (1975) calculated ^{210}Pb flux at the sediment water interface of seven Lake Michigan sites ranging from 0.14 to 0.65 $pCi/cm^2/yr$ with corresponding sedimentation rates of 17.6 and 93.8 $mg/cm^3/yr$.

Data given by Oldfield and Appleby (1984) summarize mathematically modeled ^{210}Pb chronologies of sediment cores (from a variety of locations) corroborated by other independent chronological methods and dated events. The mean ^{210}Pb flux to sediment cores calculated from these models range from 0.24 to 1.27 $pCi/cm^2/yr$.

Santschi et al. (1979) reported on the removal of the uranium and thorium series isotopes from the waters of Narragansett Bay. Removal behaviour was controlled mainly by the particulate matter concentration and the sediment resuspension rate. Settling velocities of particulate material in the bay ranged from 1.1 to greater than 28 metres per day. These settling velocities produced the following ranges of half removal times for thorium, lead, and polonium isotopes:

- ^{234}Th: 2 to 18 days;
- ^{228}Th: 14 to 64 days;
- ^{210}Pb: 10 to 64 days; and
- ^{210}Po: 27 to approximately 730 days.

The whole-lake studies of ^{226}Ra transport and accumulation conducted by Hesslein and Slavicek (1984) demonstrated that the rapid removal of this radionuclide could be modeled by a first order equation with modification for some residual value to be reached rather than assuming a continued decrease in concentration. The removal coefficient calculated for the lakes studied was reported to be in the range of 1-3.5% per day (half-times of 20-60 days).

Sedimentation rates of radionuclides depend on the sedimentation rates of sedimentary material in which (or on which) they are incorporated. Durham and Joshi (1980) found the flux of ^{210}Pb to the sediment of Lake Huron to vary from 0.15 to 0.88 $pCi/cm^2/yr$. Measurement of ^{210}Pb concentrations in the water column showed it to be essentially associated with particulate material. The settling velocity of these particles was estimated to average 7.0×10^{-4} cm/s.

Li et al. (1981) estimated the half removal time of ^{228}Th from the surface waters of the New York Bight to be 70±10 days for the slope and 20±7 days for the shallower shelf area. The most likely removal mechanism for ^{228}Th from the slope is through the phytoplankton-zooplankton-fecal pellet route. In contrast, removal from the waters of the shelf area was more likely due to adsorption of ^{228}Th onto suspended particles (organic and inorganic).

Santschi et al. (1983) examined the removal of ^{210}Pb and ^{228}Th from marine microcosms and Narragansett Bay. These radionuclides were removed from the surface waters at similar rates (1-10 days half removal times) and removal was controlled by particle settling.

The sinking rates of fecal pellets of laboratory raised zooplankton has been shown to range from 80 to 150 m/day. Sinking rates of naturally occurring fecal pellets was approximately 20% faster than the laboratory produced pellets of the same size fraction (Honjo and Roman 1978).

552 SUFFICIENCY OF INFORMATION

There is enough information concerning removal rates of ^{210}Pb, ^{226}Ra, and possibly ^{232}Th that mathematical models could be derived for the removal of these radionuclides from surface waters. However, the assumptions and the models themselves must be checked with data collected from the lakes (i.e. those impacted by uranium mining and milling) for which reliable, quantitative answers are most critical. For example, the most important assumption inherent in the dating of sediment cores via the ^{210}Pb method is that all the input of this radionuclide was deposited in the lake's sediments, with negligible amounts remaining in the water column or lost through outflow. However, Cornett et al. (1985) has demonstrated that for seven small lakes located on the Laurentian Shield,

only 16 to 80% of the total input of ^{210}Pb was transported to the lake sediments on an annual basis. Thus, studies which calculate the input of contaminants to sediments using lake sediment cores (and ^{210}Pb chronology) will underestimate the total input.

There seems to be insufficient information in the literature to provide specific mathematic models for uranium and polonium removal from surface waters.

553 RESOLUTION OF THE QUESTION

Sedimentation of the naturally occurring radionuclides in lakes is dependent upon:

- the rate of adsorption of radionuclides onto organic or inorganic particulate material;
- the seasonal availability, size and concentration of the organic and inorganic particulate material;
- the sedimentation rates of the organic and inorganic material;
- the incorporation of radionuclides into zooplankton and zooplankton fecal pellets; and
- the concentration of zooplankton.

If all the radionuclides of concern were simply scavenged by particulate material, then it would be relatively easily to relate the amount of radionuclide lost from suspension by sedimentation to the sedimentation of the particulates. Unfortunately, radium and polonium have been shown to be taken up (or have the potential to be taken up) by

phytoplankton and are thus related to the organic carbon cycle within the lake. The sedimentation of ^{210}Pb may also be influenced by the presence of organic suspended material.

Two options are available to quantify the relationship between radionuclide sedimentation and the biological and physical parameters of a lake impacted by (or potentially impacted by) uranium mining and milling.

- o whole lake studies in which sediment traps are used to quantify sedimentary material at different seasons; and
- o the use of limnocorrals as outdoor microcosms with sediment traps and the introduction of known quantities of radionuclides;

With these options, various physical and biological parameters (e.g. temperature, suspended solids, total organic carbon, phytoplankton density, zooplankton density) must be correlated with the accumulation of radionuclides in sedimentary material in a quantitative manner. Only in this way can data for sedimentation of radionuclides be derived for use in mathematical models derived from existing data or generated by these studies.

5.6 HOW DO SEDIMENTATION RATES OF RADIONUCLIDES VARY AND WHAT FACTORS INFLUENCE THE SEDIMENTATION RATES?

561 LITERATURE PERTINENT TO THE QUESTION

The variation of sedimentation rates for ^{210}Pb is given in Table 5-1.

TABLE 5-1

Sedimentation Rates of ^{210}Pb
Taken From the Published Literature

S (mm/yr)	w ($mg/cm^2/yr$)	P ($pCi/cm^2/yr$)	Reference
0.85 ± 0.03	16.7 ± 0.7	0.17	(Durham and Joshi 1981)
5.7 ± 4.9	128 + 109	0.28	
1.39 ± 0.12	29.5 ± 2.6	0.41	
2.13	44.4	0.88	(Durham and Joshi 1980)
0.25	4.8	0.15	
0.22	5.5	0.31	
0.83	17.2	0.51	
0.88	19.8	0.17	(Robbins and Edgington 1975)
0.66	12.1	0.18	
4.16	93.8	0.65	
0.78	17.6	0.14	
1.16	21.3	0.38	
0.80	14.7	0.32	
0.83	15.3	0.27	
		0.20	(Oldfield and Appleby 1984)
		0.84	
		0.59	
		0.58	
		0.72	
		0.53	
		0.46	
		1.03	
		0.64	
		0.60	
		0.46	
		1.21	
		1.33	
		0.92	
		0.17	
		0.57	

P = flux of excess ^{210}Pb to the sediment at the sediment water interface

w and S = sedimentation rates of total particulate material

The calculated variation in flux of ^{210}Pb to the sediment is probably due to a number of interactive factors which make extrapolation of rate constants measured during one season to the entire year an invalid assumption. This may be related to:

- o significant seasonal changes in organic carbon production;
- o significant seasonal changes in particulate sedimentation;
- o significant changes in lake flushing rate over the year; and
- o significant changes in ^{210}Pb input to the lake over a year.

562 SUFFICIENCY OF INFORMATION

Although there is information available concerning the variability of sedimentation rates for ^{210}Pb, there is very little information concerning the cause of the variation or how the variation might be predicated. Compared to ^{210}Pb there is little known about the variability of sedimentation rates of the other radionuclides of concern.

563 RESOLUTION OF THE QUESTION

To adequately address question 5.6, the options discussed in Section 533 must be used. Unlike Section 553, however, the variations in the biological parameters (e.g. phytoplankton density, zooplankton density, phytoplankton composition) and physical parameters (e.g. suspended inorganic particulates, temperature) and their effect on sedimentation rates must be examined to ascertain how radionuclide sedimentation rates vary with these parameters. Whole lake studies (especially those impacted by uranium mining and milling tailing) spanning an entire year

would generate much more information concerning radionuclide sedimentation variability than presently exists in the literature.

5.7 WHAT EFFECTS DO ZOOBENTHIC ORGANISMS HAVE ON THE REDISTRIBUTION OF RADIONUCLIDES WITHIN ACTIVE SEDIMENT LAYERS?

571 LITERATURE PERTINENT TO THE QUESTION

Based on ^{106}Ru profiles in 16 cores taken from Sahagawa Lake, Minnesota, mixing rates within the upper nine centimeters of sediment were calculated to average 13 cm^2/yr. Lead-210 profiles within the sediment cores were used to estimate sediment burial rates of approximately 0.4 cm/yr (Van Sickle et al. 1983). These rates were determined from a two-zone advection-diffusion model.

The work of Robbins et al. (1977, 1979) firmly established the role of benthic invertebrates such as Tubifex tubifex and Pontopereia hoyi in mixing of radionuclide contaminated sediment layers with uncontaminated sediment.

A review of the vertical mixing of lake sediments (bioturbation) was given by Fisher et al. (1980). Their laboratory microcosm study involved the migration of a ^{137}Cs labeled sediment, deposited in a marker horizon, by the activity of tubificid oligochaetes. The marker horizon was deposited at depths of 0 (i.e. surface), 2, 4 and 8 cm in separate microcosms. In the microcosms containing the marker horizon at the surface, the ^{137}Cs activity moved downward at a constant rate until it reached a depth of approximately 7 cm. Below this point, the rate decreased markedly and approached zero at a depth of approximately 8.5 cm. Labelled sediment below the surface-water interface was continuously returned to the sediment-water interface from the 6-8 cm

depth. Less labelled material was transferred to the surface sediments from depths less than 6-8 cm. The bulk of feeding activity of the oligochaetes in the laboratory microcosm (which determined the amount of material returned to sediment surface) was confined to an interval of approximately 6 to 9 cm in depth. At the termination of the experiment with marker horizon originating at the surface (120 days), the ^{137}Cs activity maximum had moved to a depth of 7.7 cm in the microcosms. With a population density of approximately 100,000 individuals/m^2, the downward velocity of the marker horizon due to the tubificid feeding was calculated to be 0.12±0.02 cm/day. This velocity corresponds to a mass flux of 25.9 g of dry sediment/cm^2/day.

In preparing a mathematical model of sediment mixing by tubificids, Fisher et al. (1980) assumed that the vertical velocity depended on the feeding rate and that the marker horizon descended uniformly (i.e. did not broaden during descent). Although, the resulting equation predicted the maximum depth to which radioactive marker horizon descended, it did not adequately predict the activity at any depth and time. To determine this parameter (i.e. activity at any depth and time) it was necessary to include in the model:

- a diffusion term to account for non-homogenous subduction;
- a more accurate description of the depth dependence of feeding rate;
- the assumption that the depth dependence of feeding was a linear function of depth for small depths, but approximated a Gaussian distribution near the zone of maximum feeding;
- the assumption that the shape of the feeding function near the zone of maximum feeding was constant in time; and

- the assumption that one-third of the sediment was not ingested by the feeding worms.

When these terms and assumptions were placed in the mathematical model, the calculated activity of the ^{137}Cs at any given depth and time was in good agreement with the experimental observations.

A review of the literature concerning depth distribution of tubificid feeding and burrowing activities by Fisher et al. (1980) implied that the depth of maximum tubificid feeding activity corresponded to the depth of peak tubificid abundance. However, calculations based on available data in the literature demonstrated that although 70% of the tubificid population was confined to the upper 3 cm of sediment, the zone of maximum feeding occurred between 2 and 7 cm. This was in general agreement with the radiotracer experiments described by Fisher et al. (1980).

The depth and rate of maximum feeding of tubificids in nature may be expected to vary with:

- the distribution of the tubificid lengths in the population;
- the species composition of the population;
- temperature;
- sediment type;
- dissolved oxygen; and
- interactions with other benthic organisms (Fisher et al. 1980).

In addition to the above factors, the density and distribution of the tubificid populations will also influence the degree of bioturbation occurring in a lake sediment.

Taken alone, the reworking of lake sediments is not particularly meaningful unless compared to local sedimentation rates. When this latter factor is determined, it may be that zoobenthic

populations of burrowing worms cause significant mixing of the surficial sediments to a depth of 6 to 8 cm.

Additional work with the ^{137}Cs marker horizon and ^{22}Na (a radionuclide weakly adorbed to sediments) demonstrated that the oligochaete Stylodrilus heringianus (at a population density of 6.7×10^4 individuals/m^2) caused a downward movement of 0.12 cm/day of the Cs-137 radiolabelled sediment to a depth of 4-6 cm. In two replicate cells with the amphipod Pontoporeia hoyi (density: $4.7 \times 10^3/m^2$) the diffusion coefficient of radiolabelled sediment was $1.43 \pm 0.25 \times 10^{-5}$ cm^2/s. No significant movement of the ^{137}Cs radiolabelled substrate was observed in laboratory microcosms containing the blood worm Chironomus tentans (density: $4.0 \times 10^3/m^2$) or the clams Pisidium sp (density $4.0 \times 10^{-3}/m^2$) and Sphaerium sp (density: $2.7 \times 10^3/m^2$). Thus, some zoobenthic organisms contribute nothing to the mixing of sediments (Krezoski and Robbins 1981).

Recent work conducted by Krezoski and Robbins (1985) involved the movement of ^{137}Cs labelled sediment layers (approximately 0.5 mm thick) placed at 10 mm intervals in a sediment column microcosm of 110 mm depth. The organism used in these studies was Stylodrilus heringianus. The authors found that the relationship between feeding distribution (labelled layer removal rate) and velocity distribution (labelled layer burial rate) for S. heringianus could be described in terms of an advection-diffusion model which accounted for depth-dependent porosity changes. The model indicated that the transfer of ^{137}Cs occurred 1.67 times faster (as the result of size-selective feeding) than mass transfer of sediment. This indicates the importance of this type of species in contaminant diagenesis.

The same type of sediment mixing by oligochaetes (primarily by the smaller oligochaete Limnodrilus hoffmeisteri) was observed by Karickhoff and Morris (1985) in their study of organic pollutant transport in bottom sediments of laboratory microcosms.

A mathematical model for particle-selective transport of tracers in sediments with conveyor-belt deposit feeders (also known as vertically oriented, deposit feeding macrobenthos) was constructed by Robbins (1985). The model provided for the quantification of the effects of particle-selective, conveyor-belt deposit feeders in a two-phase system of sediments where radionuclide reactions are limited to simple linear adsorption to sediments and radioactive decay. The data used to generate and confirm this model was supplied by laboratory microcosms. Whether or not this model will give meaningful predications in the world outside the laboratory remains to be determined.

572 SUFFICIENCY OF INFORMATION

Within the general context of sediment mixing by burrowing zoobenthic organisms, enough information is available to answer the question concerning the redistribution of radionuclides within the active sediment layer. Specifically, laboratory studies by Fisher et al. (1980) have established mathematical models, which given the correct assumptions and parameters can predict the descent of radionuclide contaminated surface sediment into the active sediment zone. This is, however, confined to sediment reworking by vertically oriented deposit feeding macrobenthos. The effects of other types of zoobenthic organisms (e.g. epibenthic macroinvertebrates) on the redistribution of radionuclides within active sediment layers is relatively unknown and would appear to be minimal.

573 RESOLUTION OF THE QUESTION

Sediment reworking by vertically oriented, deposit feeding macrobenthos results in subduction and mixing of the active sediment layer. A layer of sediment containing radionuclides above a zone of maximum feeding would move down as a reasonably discrete unit. Upon

entering the zone of maximum feeding, the downward movement of the layer abruptly slows and a substantial flux of radionuclides from that layer may be transfered to the sediment water interface. As sediment working continues, the discrete layer of radionuclide contaminated sediment is mixed throughout the active layer. The rate at which mixing occurs depends on the population density of the macrobenthos, the species composition, temperature, sediment type, dissolved oxygen, and interactions with other benthic or profunal organisms.

PART 6

REMOBILIZATION OF RADIONUCLIDES FROM SEDIMENT

PART 6 - REMOBILIZATION OF RADIONUCLIDES FROM SEDIMENT

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6.1 WHAT ARE THE POTENTIAL MECHANISMS FOR REMOBILIZATION OF RADIONUCLIDES FROM SEDIMENT, THEIR EXTENT AND OCCURRENCE?

611 LITERATURE PERTINENT TO THE QUESTION

Elsinger and Moore (1980) describe the desorption of ^{226}Ra from suspended and bottom sediments in the Pee Dee River estuary when these sediments come in contact with higher ionic strength water.

In their study of the natural radionuclides of the New York Bight, Li *et al.* (1981) hypothesize that ^{228}Th in settling particles and/or in bottom sediments had been partially regenerated to the bottom water (i.e. desorbed) during seasonal stratification and transported back to the surface water during vertical mixing. The same pattern was observed for ^{210}Pb and ^{210}Po.

In the general limnological literature, the effects of lake thermal stratification and hypolimnion oxygen depletion are known to allow the release of manganese and iron from the sediments. As both ^{210}Pb and ^{226}Ra are likely associated with the oxides of these metals in the sediment, the potential also exists for the release of radionuclides from the sediment under anoxic or near-anoxic conditions. Support for this hypothesis is given by Turekian and Cochran (1978) for the marine environment.

Hesslein and Slavicek (1984) attributed increased concentrations of ^{226}Ra in anoxic water of three experimental lakes to desorption from manganese oxides. Manganese is relatively easily reduced in natural anoxic waters and is often found in the reduced form (Mn^{+2}) in the presence of a small amount of oxygen. Thus, desorption from manganese oxides in low oxygen environments is a potential mechanism for remobilization of this radionuclide.

The general topic of remobilization of radionuclides is most often associated with problems of radioactive waste disposal. Francis (1984) briefly discusses the mobilization of toxic metals by autotrophic and heterotrophic activity. While most of this discussion is related to the creation of conditions for increased migration within a waste disposal site, some points are applicable to sediments. Several mechanisms for microbial solubilization of insoluble metals have been proposed including:

- o the production of organic acids;
- o the production of chelating agents; and
- o the metabolism of the metal-associated anion.

Microbially generated dicarboxylic acids, polyhydroxy acids, 2-ketogluctonic acid and phenolic compounds such as protocatechuic acid and salicyclic acid are effective chelating agents of heavy metals and are known to accelerate the movement of metals in soils. In many cases, a combination effect would seem to be important. For example, when the organism secretes organic acids, a dual effect may be to increase metal dissolution by lowering pH and maintenance of that metal dissolution by organic complexation. Whether or not this scenario is applicable to lake sediments is unknown. The entire subject of the significance of microbially synthesized acids and chelating agents and their effect on the mobilization of toxic metals (including radionuclides) is poorly understood.

612 SUFFICIENCY OF DATA

There is insufficient data in the scientific literature to answer the question: What are the potential mechanisms for remobilization of radionuclides from sediment, their extent and occurrence?

613 RESOLUTION OF THE QUESTION

From what information is available, the potential mechanisms for remobilization of radionuclides from sediment would appear to be related to the physical-chemical environment of the sediments. Thus, laboratory investigations of radionuclide remobilization could be conducted by exposing radionuclide contaminated sediments to various environmental variables such as:

- o deoxygenated overlying water;
- o reduced pH and redox potential of overlying water;
- o increased ionic strength of overlying water; and
- o increased pH of overlying water.

This information can subsequently be used in mathematical models. Field surveys of the lakes known to be affected by uranium mining and milling can also be made to search for the conditions which promote remobilization.

PART 7

SUMMARY

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7.1 THE RADIONUCLIDES OF CONCERN

The radionuclides which are of major interest relative to the impacts of uranium mining and milling on aquatic environments are:

- ^{226}Ra;
- ^{228}Ra (if the ore contains natural thorium);
- ^{210}Pb;
- ^{238}U and ^{234}U; and
- ^{230}Th and ^{232}Th.

7.2 THE FORM TAKEN BY RADIONUCLIDES ENTERING THE AQUATIC ENVIRONMENT

There has been, to date, no definitive study of the form taken by radionuclides entering the aquatic environment from uranium milling tailings. Based on the conditions within the tailings (i.e. pH, redox potential, mineralogy of the tailings), a knowledge of the aqueous chemistry of the radionuclides and information concerning the chemical characteristics of the groundwater or surface water flowing through (or over) the tailings, certain assumptions can be made concerning the expected form of the radionuclides. However, because there are so many variables related to the final form taken by the radionuclide, each site must be dealt with on a case-by-case basis.

It is reasonable to assume from the published literature that the organic species involved in formation of a radionuclide-organic complex would be humic substances. This material constitutes the bulk of organic matter in natural waters and is known to form organometallic complexes with a variety of metals.

7.3 REMOVAL OF RADIONUCLIDES FROM SURFACE WATERS

The general mechanisms by which metals (and radionuclides) are removed from surface waters can be described as:

- o chemical adsorption to organic and inorganic particulate material;
- o absorption by living particulate material;
- o incorporation into zooplankton fecal pellets;
- o precipitation scavenging; and
- o exchange.

From the information available in the literature, chemical adsorption by living and nonliving particulate matter appears to play the largest role in removing radionuclides from surface waters by sedimentation. A combination of precipitation scavenging and adsorption may be important for thorium, but insufficient information is available to clearly establish this. Very little information is available on the occurrence of exchange (i.e. cation or anion exchange of the ionic or complexed ionic form of the radionuclides). As well, little information is available on the role played by organic detritus in radionuclide removal.

Biological organisms can remove radionuclides from surface waters by several mechanisms:

- o adsorption to the exoskeleton or cell wall of living organisms and their subsequent sedimentation at death;
- o absorption into the organism with transfer to the sediment at the organisms death as part of the organic detritus;
- o consumption of contaminated detritus or living cells and release in a fecal pellet with subsequent sedimentation; and

- absorption through the gut and sequestering in the organism body.

Of the three basic zones in a lake (i.e. Limnetic or pelagic, profundal, and littoral) the littoral zone has the potential to have the widest diversity of life available for the removal of radionuclides from water. In particular, aquatic vascular plants, attached macro and microalgae (normal residents of the littoral zone) appear to be extremely efficient in the removal of at least some radionuclides from the water.

Removal of radionuclides by organisms in the limnetic zone is usually related to the diversity and concentration of only two groups (i.e. the phytoplankton and zooplankton). The profundal zone may contain some organisms normally associated with the limnetic zone as well as benthic and epibenthic organisms which enter the water column at certain times during a diurnal cycle. It is not known how much these organisms contribute to the removal of radionuclides, but based on their relative density (compared to organisms in the other two zones) they probably contribute little.

7.4 SEDIMENTATION OF RADIONUCLIDES AND DISPERSION WITHIN SEDIMENTS

There is virtually nothing known about the sedimentary history of lakes known to be affected by uranium milling operations. Whether sediment cores collected from these areas can be used to define a sedimentary history useful for predictive modelling can only be determined by field studies at these sites. Mathematical models are available for use in determining sediment accumulation from single core measurements and their transformation to basin-wide influx rates.

The published literature can give details of specific techniques and equipment to be used for sedimentary history reconstruction, but specific locations for sediment core collections must be mapped out on a lake-by-lake basis.

Once sediment cores are taken, the establishment of a time-depth relationship must be established by means other than those utilizing natural radionuclides (i.e. ^{210}Pb, ^{230}Th, ^{232}Th). Carbon-14, palynology, or ^{137}Cs may be candidates for this analysis.

The use of ^{210}Pb to establish geochronologies of lake and marine sediments has lead to a great deal of published information concerning the sedimentation rates of this particular radionuclide (as well as the total sedimentary mass per time). Mathematically modelled ^{210}Pb chronologies of sediment cores from a variety of locations yielded ^{210}Pb flux to the sediment ranging from 0.24 to 1.27 $pCi/cm^2/yr$.

The calculated variation in flux of ^{210}Pb to the sediment is probably due to a number of interactive factors in the lakes related to

- changes in organic carbon production;
- changes in particulate sedimentation;
- changes in lake flushing; and
- changes in ^{210}Pb input.

Studies have been conducted which establish the role of vertically oriented, deposit feeding macrobenthos in the redistribution of radionuclides (and other pollutants) within the active sediment layer. The most important of these studies used populations of tubificid (oligochaete) worms to demonstrate the downward movement of a radionuclide contaminated sediment layer. The data from this study was used to develop a mathematical model to predict the activity of the radionuclide at any depth and time. The critical component of this model is the depth and

rate of maximum feeding of the tubificids in real world situations. This component may be expected to vary with:

- o the distribution of the lengths of tubificids in the population;
- o the species composition of the population;
- o temperature;
- o sediment type;
- o dissolved oxygen; and
- o interactions with other benthic organisms.

In addition to the above factors, the density and distribution of tubificid populations will also influence the degree of bioturbation occuring in the lake sediment.

7.5 REMOBILIZATION OF RADIONUCLIDES

No studies were found during the literature survey which involved remobilization of radionuclides from sediment. Remobilization or release of radionuclides from particulate material or sediments under anoxic or partially anoxic conditions has been hypothesized to explain increases of specific dissolved radionuclides in the water column.

7.6 INFORMATION NEEDS

To adequately address the questions related to the sedimentation of radionuclides in the natural environment which have not been adequately addressed by the published literature, various laboratory and field investigations must be conducted.

The form (ionic versus organic complex) of the radionuclides entering the aquatic environment must be determined by the collection of water samples (both surface and groundwater) and the separation of organically complexed and ionic radionuclides. It is anticipated that seasonal variation might occur in the relative preparation of these two fractions. Thus, seasonal collection of water samples will be necessary to establish variability.

Although radionuclides are known to be removed from surface waters by a variety of mechanisms, the specific local environment of a lake known to be influenced by uranium tailings will have a specific combination of these mechanisms. These particular environments must be studied to identify and quantify each interacting mechanism. Seasonal variations in phyto- and zooplankton populations must be related to sedimentation and the radionuclide content of the sedimentary material. The biomass and radionuclide concentrations of littoral aquatic vegetation and its input into the total sedimentary material available to the lake must also be quantitatively determined. The various types of suspended material in these lakes must be examined in the laboratory for organic content, surface adsorption features, ion exchange capacity, and settling velocity. This will ensure a quantitative evaluation of radionuclide sedimentation rates.

The sedimentary history of lakes influenced by uranium tailings should be examined to determine if:

- historical patterns of sediment deposition can be determined from sediment cores; and
- if that data obtained from sediment cores can be used in existing models of sediment deposition.

The sediment coring program should be part of a wider program to quantify sediment inputs and outputs in the lakes as well as the determination of the extent of sedimentation and sediment movement in various portions of the lake basin.

The effect of vertically oriented, deposit feeding macrobenthos on sediment mixing has been quantified. Whether the models developed for this mechanism can be applied to lakes influenced by uranium tailings depends on a quantitative and taxonomic evaluation of the benthos inhabiting the lake. This information will also determine the extent and occurrence of mixing.

PART 8

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APPENDIX A

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